

JC10 Rec'd PCT/PTO 25 FEB 2002

FORM PTO-1390 (REV. 9-2001)		U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>		ATTORNEY'S DOCKET NUMBER <b>566.41244X00 filed February 25, 2002</b>	
		U S APPLICATION NO. (If known, see 37 CFR 1.5) <b>107069273</b>	
INTERNATIONAL APPLICATION NO <b>PCT/JP00/05766</b>	INTERNATIONAL FILING DATE <b>August 25, 2000</b>	PRIORITY DATE CLAIMED <b>August 25, 1999</b>	
<b>TITLE OF INVENTION</b> <b>ADHESIVE, METHOD OF CONNECTING WIRING TERMINALS AND WIRING STRUCTURE</b>			
APPLICANT(S) FOR DO/EO/US <b>ARIFUKU, MOTOHIRO WATANABE, ITSUO MOTOMURA, KOUJI KOBAYASHI, KOUJI GOTO, YASUSHI</b>			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</p> <p>4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) a. <input type="checkbox"/> is transmitted hereto (required only if not communicated by the International Bureau). b. <input checked="" type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office(RO/US)</p> <p>6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p>			
<b>Items 11 to 20 below concern document(s) or information included:</b>			
<p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.</p> <p>14. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input checked="" type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</p> <p>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</p> <p>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</p> <p>20. <input checked="" type="checkbox"/> Other items or information: <b>Figs. 1a-1d,2a-2b; Credit Card Payment Form</b></p>			

U.S. APPLICATION NO. (Unknown, see 37 CFR 1.492(e)) <b>107069273</b>	INTERNATIONAL APPLICATION NO <b>PCT/JP00/05766</b>	ATTORNEY'S DOCKET NUMBER <b>566.41244X00</b>		
21. The following fees are submitted		<b>CALCULATIONS PTO USE ONLY</b>		
<b>BASIC NATIONAL FEE (37 CFR 1.492(a) (1) - (5)):</b>				
<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO .....		\$1040.00		
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO .....		\$890.00		
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO .....		\$740.00		
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) .....		\$710.00		
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) .....		\$100.00		
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>		<b>\$890.00</b>		
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492(e)).		<input type="checkbox"/> 20 <input type="checkbox"/> 30 \$		
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total Claims	24 - 20 =	4	x \$18.00	\$72.00
Independent Claims	2 - 3 =	0	x \$84.00	\$
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)			+ \$280.00	\$
<b>TOTAL OF ABOVE CALCULATIONS =</b>			<b>\$962.00</b>	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by $\frac{1}{2}$ . +			\$	
<b>SUBTOTAL =</b>			<b>\$962.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492(f)).			<input type="checkbox"/> 20 <input type="checkbox"/> 30 \$	
<b>TOTAL NATIONAL FEE =</b>			<b>\$962.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +			\$	
<b>TOTAL FEES ENCLOSED =</b>			<b>\$962.00</b>	
			<b>Amount to be refunded:</b>	\$
			<b>charged:</b>	\$
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the fees is enclosed				
b. <input type="checkbox"/> Please charge my Deposit Account No <b>01-2135</b> in the amount of \$ _____ to cover the above fees A duplicate copy of this sheet is enclosed				
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No <b>01-2135</b> . A duplicate copy of this sheet is enclosed				
d. <input checked="" type="checkbox"/> Fees are to be charged to a credit card. <b>WARNING:</b> Information on this form may become public. <b>Credit card information should not be included on this form.</b> Provide credit card information and authorization on PTO-2038.				
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>				
SEND ALL CORRESPONDENCE TO				
<b>Antonelli, Terry, Stout &amp; Kraus, LLP</b> <b>1300 North Seventeenth Street</b> <b>Suite 1800</b> <b>Arlington, VA 22209</b> <b>USA</b>				
 <b>SIGNATURE</b> <b>Carl I. Brundidge</b> <b>NAME</b> <b>29,621</b> <b>REGISTRATION NO.</b>				

566.41244X00

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): ARUFUKU, et al.

Filed: February 25, 2002

Serial No.: Not yet assigned

For: ADHESIVE, METHOD OF CONNECTING WIRING TERMINALS  
AND WIRING STRUCTURE

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231

February 25, 2002

Sir:

Please amend the above-identified application, prior to  
examination thereof, as follows:

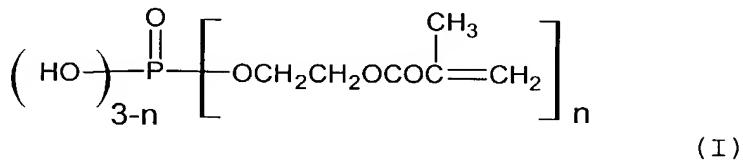
IN THE SPECIFICATION

Replace the paragraph beginning on page 9, line 23 with the  
following:

Peroxydicarbonates preferable for the present invention  
may include di(n-propyl) peroxydicarbonate, di(isopropyl)  
peroxydicarbonate, bis (4-t-butylcyclohexyl)  
peroxydicarbonate, di(2-ethoxyethyl) peroxydicarbonate,  
di(2-ethylhexyl) peroxydicarbonate, di(methoxybutyl)  
peroxydicarbonate, di(3-methyl-3-methoxybutyl)  
peroxydicarbonate and the like.

Replace the paragraph beginning on page 13, line 13 with the  
following:

The radically polymerizable substance described above may also be used in combination with a radically polymerizable substance having a phosphoric ester structure represented by the following chemical formula (I). This is preferable because the bond strength on the surface of inorganic matter such as metal is improved.



wherein n is 1, 2 or 3.

Replace the paragraph on page 24, line 17 with the following:

As a silane coupling agent having an amino group, it may include, e.g.,

N- $\beta$ -(aminoethyl)- $\gamma$ -aminopropyltrimethoxysilane,

N- $\beta$ -(aminoethyl)- $\gamma$ -aminopropylmethyldimethoxysilane,  $\gamma$ -aminopropyltriethoxysilane, N-phenyl- $\gamma$ -aminopropyltrimethoxysilane and the like.

Replace the paragraph beginning on page 29, line 18 with the following:

The urethane acrylate A obtained in the step (1), a phosphate type acrylate (available from Kyoeisha Chemical Co., Ltd.; tradename: P2M), the silicone particles and t-hexyl peroxy-2-ethylhexanone (a free radical generator) were so

mixed as to be in amounts of 99 g, 1 g, 30 g and 5 g, respectively, in solid weight ratio, and 3% by volume of the conductive particles obtained in the step (3) were dispersed to obtain a liquid adhesive.

Please replace the paragraph beginning on page 31, line 17 with the following:

First, on the surface of a glass substrate 11 of 1.1 mm thick, wirings 12 of indium-tin oxide (ITO) were formed by vacuum deposition to prepare an ITO substrate member (surface resistivity: <20  $\Omega$ /square) 10 [Fig. 1 (a)]. On its side where the wiring 12 was formed, an adhesive layer 15 formed of an adhesive 13 containing conductive particles 14 (the one prepared in each Example and Comparative Example) was formed [Fig. 1 (b)].

IN THE CLAIMS

Please amend Claims 5, 7, 9, and 11 - 13 as follows:

5. (amended) The wiring-terminal-connecting adhesive according to claim 3, wherein said silicone particles are contained in an amount of from 5 parts by weight to 200 parts by weight based on 100 parts by weight of the total of said radically polymerizable substance and said film-forming material.

7. (amended) The wiring-terminal-connecting adhesive according to claim 1, which further comprises conductive particles.

9. (amended) A method of connecting wiring terminals, comprising interconnecting wiring terminals electrically with the wiring-terminal-connecting adhesive according to claim 1, wherein at least two wiring members have the connecting terminal individually.

11. (amended) The method of connecting wiring terminals according to claim 9, wherein at least one of said wiring members has

a substrate comprising at least one of an insulating organic material and glass.

12. (amended) The method of connecting wiring terminals according to claim 9, wherein at least one of said wiring members has

at least one selected from silicon nitride, silicone resin and polyimide resin, at its surface.

13. (amended) A wiring structure comprising at least two wiring members which have a wiring terminal individually, wherein

the wiring terminals of said wiring members are

electrically interconnected with the wiring-terminal-connecting adhesive according to claim 1.

Please add the following new claim to the application:

--14. A method of connecting wiring terminals, comprising interconnecting wiring terminals electrically with the wiring-terminal-connecting adhesive according to claim 8, wherein at least two wiring members have the connecting terminal individually.

15. The method of connecting wiring terminals according to claim 14, wherein at least one of said connecting terminals has a surface which is formed of at least one selected from gold, silver, tin, a platinum group metal and indium-tin oxide.

16. The method of connecting wiring terminals according to claim 14, wherein at least one of said wiring members has a substrate comprising at least one of an insulating organic material and glass.

17. The method of connecting wiring terminals according to claim 14, wherein at least one of said wiring members has at least one selected from silicon nitride, silicone resin and polyimide resin, at its surface.

18. The wiring-terminal-connecting adhesive according to claim 4, wherein said silicone particles are contained in an amount of from 5 parts by weight to 200 parts by weight based on 100 parts by weight of the total of said radically polymerizable substance and said film-forming material.

19. The wiring-terminal-connecting adhesive according to claim 2, which further comprises conductive particles.

20. The wiring-terminal-connecting adhesive according to claim 3, which further comprises conductive particles.

21. The wiring-terminal-connecting adhesive according to claim 4, which further comprises conductive particles.

22. A wiring structure comprising at least two wiring members which have a connecting terminal individually, wherein the connecting terminals of said wiring members are electrically interconnected with the wiring-terminal-connecting adhesive according to claim 2.

23. A wiring structure comprising at least two wiring members which have a connecting terminal individually, wherein the connecting terminals of said wiring members are electrically interconnected with the wiring-terminal-connecting adhesive according to claim 3.

24. A wiring structure comprising at least two wiring members which have a connecting terminal individually, wherein the connecting terminals of said wiring members are electrically interconnected with the wiring-terminal-connecting adhesive according to claim 4.--

REMARKS

The present application is a National Phase filing of PCT/JP00/05766. The claims were previously amended on March 26, 2001 and July 23, 2001 under PCT Article 34. Claims 5, 7, 9, and 11 - 13 are further amended herein, and new Claims 14 - 25 are added, in order to eliminate multiple dependent claims. Claims 9 and 13, and portions of the specification, are amended to correct minor errors. It is respectfully requested that these amendments be entered prior to calculating the filing fee in connection with the above-identified application.

In view of all of the foregoing, entry of the present amendments, and examination of the above-identified application in due course, are respectfully requested.

Kindly charge any additional fees due, or credit overpayment of fees, to Deposit Account No. 01-2135. (File No. 566.41244X00).

Respectfully submitted,

ANTONELLI, TERRY, STOUT & KRAUS, LLP

  
Carl I. Brundidge  
Registration No. 29,621

1300 North Seventeenth Street  
Suite 1800  
Arlington, VA 22209  
Tel.: 703-312-6600  
Fax.: 703-312-6666

CIB/RTW:lcb

Marked up copy to show changes made

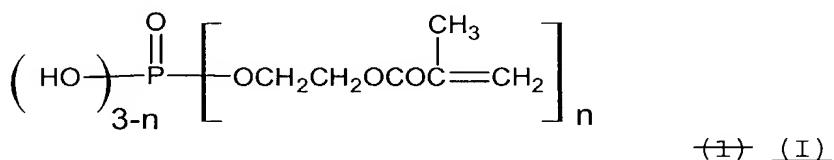
IN THE SPECIFICATION

Replace the paragraph beginning on page 9, line 23 with the following:

Peroxydicarbonates preferable for the present invention may include di(n-propyl) peroxydicarbonate, di(isopropyl) peroxydicarbonate, bis (4-t-butylcyclohexyl) peroxydicarbonate, di(2-ethoxyethyl) peroxydicarbonate, di(2-ethylhexyl) peroxydicarbonate, di(methoxybutyl) peroxydicarbonate, di(3-methyl-3methoxybutyl) di(3-methyl-3-  
methoxybutyl) peroxydicarbonate and the like.

Replace the paragraph beginning on page 13, line 13 with the following:

The radically polymerizable substance described above may also be used in combination with a radically polymerizable substance having a phosphoric ester structure represented by the following chemical formula (I). This is preferable because the bond strength on the surface of inorganic matter such as metal is improved.



wherein n is 1, 2 or 3.

Replace the paragraph on page 24, line 17 with the following:

As a silane coupling agent having an amino group, it may include, e.g.,

N-β-(aminoethyl)γ-aminopropyltrimethoxysilane, N-β-(aminoethyl)-γ-aminopropyltrimethoxysilane, N-β-(aminoethyl)γ-aminopropylmethyldimethoxysilane, N-β-(aminoethyl)-γ-aminopropylmethyldimethoxysilane, γ-aminopropyltriethoxysilane, N-phenyl-γ-aminopropyltrimethoxysilane and the like.

Replace the paragraph beginning on page 29, line 18 with the following:

The urethane acrylate A obtained in the step (1), a phosphate type acrylate (available from Kyoeishayushi Kabushiki Kaiya Kyoeisha Chemical Co., Ltd.; tradename: P2M), the silicone particles and t-hexyl peroxy-2-ethylhexanone (a free radical generator) were so mixed as to be in amounts of 99 g, 1 g, 30 g and 5 g, respectively, in solid weight ratio, and 3% by volume of the conductive particles obtained in the step (3) were dispersed to obtain a liquid adhesive.

Please replace the paragraph beginning on page 31, line 17 with the following:

First, on the surface of a glass substrate 11 of 1.1 mm thick, wirings 12 of indium-tin oxide (ITO) were formed by

vacuum deposition to prepare an ITO substrate member (surface resistivity: <20  $\Omega$ /square) 10 [Fig. 1 (a)]. On its side where the wiring 12 was formed, an adhesive layer 15 formed of an adhesive 13 containing conductive particles 14 (the one prepared in each Example and Comparative Example) was formed [Fig. 1 (b)].

IN THE CLAIMS

5. (amended) The wiring-terminal-connecting adhesive according to claim 3 ~~or 4~~, wherein said silicone particles are contained in an amount of from 5 parts by weight to 200 parts by weight based on 100 parts by weight of the total of said radically polymerizable substance and said film-forming material.

7. (amended) The wiring-terminal-connecting adhesive according to ~~any one of claims 1 to 5~~ claim 1, which further comprises conductive particles.

9. (amended) A method of connecting wiring terminals, comprising interconnecting ~~connecting~~ wiring terminals electrically with the wiring-terminal-connecting adhesive according to ~~any one of claims 1 to 5, 7 and 8~~ claim 1, wherein at least two wiring members have the connecting terminal individually.

11. (amended) The method of connecting wiring terminals according to claim 9 ~~or 10~~, wherein at least one of said wiring members has

a substrate comprising at least one of an insulating organic material and glass.

12. (amended) The method of connecting wiring terminals according to ~~any one of claims 9 to 11~~ claim 9, wherein at least one of said wiring members has

at least one selected from silicon nitride, silicone resin and polyimide resin, at its surface.

13. (amended) A wiring structure comprising at least two wiring members which have a connecting wiring terminal individually, wherein

the connecting wiring terminals of said wiring members are electrically interconnected with the wiring-terminal-connecting adhesive according to ~~any one of claims 1 to 5, 7 and 8~~ claim 1.

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## SPECIFICATION

### ADHESIVE, METHOD OF CONNECTING WIRING TERMINALS

AND

5

### WIRING STRUCTURE

#### TECHNICAL FIELD

This invention relates to a wiring-connecting adhesive, and a wiring-terminal-connecting method and 10 a wiring structure which make use of such an adhesive.

#### BACKGROUND ART

In recent years, wiring density is being made higher in the field of precision electronic machinery, so that 15 electrodes are formed in very small width and distance.

Hence, there is a possibility that the wiring may fall off, come off or come to misregistration. To solve this problem, adhesive compositions for electric and electronic use which have a good low-temperature 20 curability and having a pot life have been developed (e.g., Japanese Patent Application Laid-open No. 11-97825).

However, conventional wiring and connecting members have had a problem that they differ in bond strength

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depending on the type of materials constituting the wiring to be connected. In particular, where the substrate which supports wiring terminals is made of an insulating organic material such as polyimide resin, or glass, or where 5 silicon nitride, silicone resin or polyimide resin is coated or stays attached at the surface of a wiring member, there has been the problem of a very low bond strength.

#### DISCLOSURE OF THE INVENTION

10 An object of the present invention is to provide an adhesive which is suited for electric and electronic use and can achieve a high bond strength especially even when it bonds a wiring member in which the substrate which supports wiring terminals is made of an insulating 15 organic matter or glass, or a wiring member having silicon nitride, silicone resin and/or polyimide resin at its surface at least in part, and provide a wiring-terminal-connecting method and a wiring structure which make use of such an adhesive.

20 The present invention provides a wiring-terminal-connecting adhesive comprising (1) a curing agent capable of generating a free radical upon heating, (2) a radically polymerizable substance and (3) silicone particles. The adhesive of the present

invention can be used to connect terminals electrically by interposing the adhesive between wiring substrates which are so disposed that wiring terminals provided on their surfaces face to one another, and heating these wiring substrates under application of a pressure. In the present invention, the terminals may also be electrodes.

The adhesive of the present invention may further comprise (4) a film-forming material. As the 10 film-forming material, phenoxy resin is preferred.

The adhesive of the present invention may still further comprise (5) conductive particles. The conductive particles may preferably be composed of at least one of gold, silver and a platinum group metal at least at their surfaces.

The silicone particles in the adhesive of the present invention may preferably be contained in an amount of from 5 to 200 parts by weight based on 100 parts by weight of the radically polymerizable substance (when the 20 film-forming material is contained, based on 100 parts by weight of the total of the radically polymerizable substance and the film-forming material).

The silicone particles used in the adhesive of the present invention may also preferably have a modulus

of elasticity of from 0.1 to 100 MPa at 25°C (room temperature).

The present invention also provides a wiring-terminal-connecting adhesive film comprising a 5 first layer formed of a composition containing (1) a curing agent capable of generating a free radical upon heating, (2) a radically polymerizable substance and (3) silicone particles; and a second layer formed of a composition containing (5) conductive particles, (2) a radically polymerizable substance and (3) silicone 10 particles, wherein the first and second layers are formed in layers.

The present invention still also provides a method of connecting wiring terminals, comprising 15 interconnecting connecting terminals electrically with the adhesive of the present invention, wherein at least two wiring members have the connecting terminal individually.

The connecting method of the present invention is a method in which a first wiring member having a first connecting terminal and a second wiring member having a second connecting terminal which are so disposed that the terminals face to each other in the state the adhesive of the present invention is interposed between them are

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heated under application of a pressure in the direction of bonding to connect the first connecting terminal and the second connecting terminal electrically.

The connecting method of the present invention is especially suited when at least one of the connecting terminals has a surface which is formed of at least one of gold, silver, tin, a platinum group metal and/or indium-tin oxide (ITO). The connecting method of the present invention is also suited when at least one of the wiring members has a substrate comprising an insulating organic material and/or glass. Moreover, the connecting method of the present invention can achieve a superior bond strength even when at least one of the wiring members has at least one of silicon nitride, silicone resin and polyimide resin at its surface.

The present invention further provides a wiring structure comprising at least two wiring members which have a connecting terminal individually, wherein the connecting terminals of the wiring members are electrically interconnected with the adhesive of the present invention.

The wiring structure of the present invention has a structure in which a first wiring member having a first connecting terminal and a second wiring member having

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a second connecting terminal are so disposed that the first connecting terminal and the second connecting terminal face to each other in the state the adhesive of the present invention is interposed between them, 5 and the first connecting terminal and the second connecting terminal are electrically connected.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figs. 1 and 2 are illustrations showing the steps 10 of connecting wiring terminals.

#### BEST MODES FOR PRACTICING THE INVENTION

(1) Curing agent capable of generating a free radical upon heating:

15 The curing agent capable of generating a free radical upon heating, contained in the adhesive of the present invention, is a substance which undergoes decomposition upon heating to generate a free radical, as exemplified by a peroxide or an azo compound. This curing agent may 20 appropriately be selected taking account of the intended connecting temperature, connecting time, pot life and so forth. In view of the highness of reactivity and the length of pot life, the curing agent may preferably be an organic peroxide having a temperature of 40°C or above

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for the 10-hour half-life and a temperature of 180°C or below for the 1-minute half-life, and more preferably an organic peroxide having a temperature of 60°C or above for the 10-hour half-life and a temperature of 170°C or 5 below for the 1-minute half-life.

When connected in a time of 10 seconds or less, the curing agent may preferably be mixed in an amount of from 0.1 to 30 parts by weight, and more preferably from 1 to 20 parts by weight, based on 100 parts by weight 10 of the radically polymerizable substance (when the film-forming material is contained, based on 100 parts by weight of the total of the radically polymerizable substance and the film-forming material) in order to achieve a sufficient rate of reaction. If the curing 15 agent is mixed in an amount of less than 0.1 part by weight, any sufficient rate of reaction can not be achieved to tend to make it difficult to achieve a good bond strength and a low connection resistance. If it is mixed in an amount of more than 30 parts by weight, the adhesive 20 may have a low flowability or a high connection resistance, and the adhesive tends to have a short lifetime.

Curing agents preferable for the present invention may include diacyl peroxides, peroxydicarbonates, peroxy esters, peroxy ketals, dialkyl peroxides, hydroperoxides

and silyl peroxides.

In order to keep the connecting terminals of the wiring member from corroding, any chloride ion or organic acid contained in the curing agent may preferably be 5 not more than 5,000 ppm. Those in which any organic acid generated after thermal decomposition can be in a small content are more preferred. Stated specifically, peroxy esters, dialkyl peroxides, hydroperoxides, silyl peroxides, and the like are preferred. In particular, 10 it is preferable to select the curing agent from peroxy esters, which can achieve a high reactivity. Of these curing agents, any one compound may be used alone, or two or more compounds may be used in appropriate combination.

15 Peroxy esters preferable for the present invention may include cumyl peroxyneodecanoate, 1,1,3,3-tetramethylbutyl peroxyneodecanoate, 1-cyclohexyl-1-methylethyl peroxyneodecanoate, t-hexyl peroxyneodecanoate, t-butyl peroxypropionate, 20 1,1,3,3-tetramethylbutyl peroxy-2-ethylhexanone, 2,5-dimethyl-2,5-di(2-ethylhexanoylperoxy)hexane, 1-cyclohexyl-1-methylethyl peroxy-2-ethylhexanone, t-hexyl peroxy-2-ethylhexanone, t-butyl peroxy-2-ethylhexanone, t-butyl peroxyisobutyrate,

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1,1-bis(t-butyloxy)cyclohexane, t-hexyl

peroxyisopropylmonocarbonate, t- butyl

peroxy-3,5,5-trimethylhexanone, t-butyl

peroxylaurate,

5 2,5-dimethyl-2,5-di(m-toluoylperoxy)hexane, t-butyl

peroxyisopropylmonocarbonate, t-butyl

peroxy-2-ethylhexylmonocarbonate, t-hexyl

peroxybenzoate, t-butyl peroxyacetate and the like.

Dialkyl peroxides preferable for the present

10 invention may include

$\alpha, \alpha'$ -bis(t-butyloxy)diisopropylbenzene, dicumyl

peroxide, 2,5-dimethyl-2,5-di(t-butyloxy)hexane,

t-butyl cumyl peroxide and the like.

Hydroperoxides preferable for the present invention

15 may include diisopropylbenzene hydroperoxide, cumene  
hydroperoxide, and the like.

Diacyl peroxides preferable for the present invention  
may include isobutyl peroxide, 2,4-dichlorobenzoyl  
peroxide, 3,5,5-trimethylhexanoyl peroxide, octanoyl  
20 peroxide, lauroyl peroxide, stearoyl peroxide, succinic  
peroxide, benzoyl peroxytoluene, benzoyl peroxide and  
the like.

Peroxydicarbonates preferable for the present  
invention may include di(n-propyl) peroxydicarbonate,

di(isopropyl) peroxydicarbonate,  
bis(4-t-butylcyclohexyl) peroxydicarbonate,  
di(2-ethoxyethyl) peroxydicarbonate, di(2-ethylhexyl)  
peroxydicarbonate, di(methoxybutyl) peroxydicarbonate,  
5 di(3-methyl-3methoxybutyl) peroxydicarbonate and the  
like.

Peroxy ketals may include

1,1-bis(t-hexylperoxy)-3,3,5-trimethylcyclohexane,  
1,1-bis(t-hexylperoxy)cyclohexane,  
10 1,1-bis(t-butylperoxy)-3,3,5-trimethylcyclohexane,  
1,1-bis(t-butylperoxy)cyclododecane,  
2,2-bis(t-butylperoxy)decane and the like.

Silyl peroxides may include t-butyltrimethylsilyl  
peroxide, bis(t-butyl)dimethylsilyl peroxide,  
15 t-butyltrivinylsilyl peroxide,  
bis(t-butyl)divinylsilyl peroxide,  
tris(t-butyl)vinylsilyl peroxide, t-butyltriallylsilyl  
peroxide, bis(t-butyl)diallylsilyl peroxide,  
tris(t-butyl)allylsilyl peroxide and the like.

20 Of these curing agents capable of generating free  
radicals upon heating, any one compound may be used alone,  
or two or more compounds may be used in combination.  
Also, any of these curing agents may be used in combination  
with a decomposition accelerator, an inhibitor and so

forth.

These curing agents may be coated with a polyurethane type or polyester type polymeric compound so as to be made into microcapsules. Such curing agents are also 5 preferable for the present invention because the pot life can be made longer.

(2) Radically polymerizable substance:

The radically polymerizable substance to be contained in the adhesive of the present invention is a substance 10 having a functional group which can polymerize by the action of radicals. This radically polymerizable substance may include acrylates, methacrylates, maleimide compounds and the like, which may be in the state of either of monomers and oligomers. A monomer 15 and an oligomer may also be used in combination.

As specific examples of acrylates and methacrylates preferable for the adhesive of the present invention, they may include methyl acrylate, ethyl acrylate, isopropyl acrylate, isobutyl acrylate, ethylene glycol 20 diacrylate, diethylene glycol diacrylate, trimethylolpropane triacrylate, tetramethylolmethane tetraacrylate, 2-hydroxy-1,3-diacryloxypropane, 2,2-bis[4-(acryloxymethoxy)phenyl]propane, 2,2-bis[4-(acryloxypolyethoxy)phenyl]propane,

dicyclopentenyl acrylate, tricyclodecanyl acrylate, tris(acryloyloxyethyl) isocyanurate, urethane acrylate, etc., and methacrylates corresponding to these.

Maleimide compounds preferable for the adhesive of the present invention are those having at least two maleimide groups in the molecule. Such maleimide compounds may include, e.g.,

1-methyl-2,4-bismaleimidobenzene,  
N,N'-m-phenylenebismaleimide,  
10 N,N'-p-phenylenebismaleimide,  
N,N'-m-toluylenebismaleimide,  
N,N'-4,4-biphenylenebismaleimide,  
N,N'-4,4-(3,3'-dimethyl-biphenylene)bismaleimide,  
N,N'-4,4-(3,3'-dimethyldiphenylmethane)bismaleimide,  
15 N,N'-4,4-(3,3'-diethyldiphenylmethane)bismaleimide,  
N,N'-4,4-diphenylmethanebismaleimide,  
N,N'-4,4-diphenylpropanebismaleimide,  
N,N'-3,3'-diphenylsulfonebismaleimide,  
N,N'-4,4-diphenyl ether bismaleimide,  
20 2,2-bis{4-(4-maleimidophenoxy)phenyl}propane,  
2,2-bis{3-s-butyl-4,8-(4-maleimidophenoxy)phenyl}propane, 1,1-bis{4-(4-maleimidophenoxy)phenyl}decane,  
4,4'-cyclohexylidene-bis{1-(4-maleimidophenoxy)phenyl  
}-2-cyclohexylbenzene,

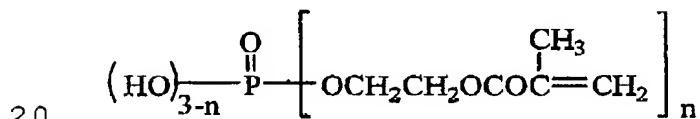
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2,2-bis(4-(4-maleimidophenoxy)phenyl)hexafluoropropane and the like

Of these radically polymerizable substances, compounds having at least one of a dicyclopentenyl group, 5 a tricyclodecanyl group and a triazine ring are preferred because adhesive cured products can be improved in heat resistance.

Of these radically polymerizable substances, any one may be used alone or two or more may be used in 10 combination. If necessary, a polymerization inhibitor such as hydroquinones or methyl ether hydroquinones may appropriately be used.

The radically polymerizable substance described above may also be used in combination with a radically 15 polymerizable substance having a phosphoric ester structure represented by the following chemical formula (I). This is preferable because the bond strength on the surface of inorganic matter such as metal is improved.



(1)

wherein n is 1, 2 or 3.

Such a radically polymerizable substance having a phosphoric ester structure can be obtained by allowing phosphoric anhydride to react with 2-hydroxyethyl acrylate or methacrylate. It may specifically include 5 mono(2-methacryloyloxyethyl) acid phosphate, di(2-methacryloyloxyethyl) acid phosphate and the like. Of these, any one compound may be used alone, or two or more may be used in combination.

The radically polymerizable substance having a phosphoric ester structure may preferably be mixed in an amount of from 0.01 to 50 parts by weight, and more preferably from 0.5 to 5 parts by weight, based on 100 parts by weight of the total weight of the radically polymerizable substance(s) (when the film-forming 15 material is contained, based on 100 parts by weight of the total of the radically polymerizable substance(s) and the film-forming material). If it is less than 0.01 part by weight, the bond strength to the surface of inorganic matter such as metal may be improved with 20 difficulty. If it is more than 50 parts by weight, the expected curing performance may not be achieved.

(3) Silicone particles:

The silicone particles may be obtained by a method in which, e.g., a silane compound (such as a

methyltrialkoxysilane or a partial hydrolysis-condensation product thereof) is added to an aqueous alcohol solution the pH of which has been adjusted with sodium hydroxide or ammonia to more than 5 and the mixture formed is hydrolyzed to effect polycondensation, or may be obtained by, e.g., copolymerization of an organosiloxane.

In the adhesive of the present invention, silicone particles having a functional group such as a hydroxyl group, an epoxy group, a ketimine, a carboxyl group or a mercapto group at the molecular terminal or on the side chain in the molecule are preferred because the dispersibility in the film-forming material and radically polymerizable substance(s) can be improved.

15 In the present invention, the incorporation of the silicone particles in the adhesive enables achievement of a very high bond strength even on the wiring member in which the substrate which supports connecting terminals is made of an insulating organic matter or glass, or 20 on the wiring member having silicon nitride, silicone compound or polyimide resin at its surface. Also, in the case when the adhesive is made into a film by the use of the film-forming material, the film is improved in its releasability from a base material, and hence

is improved in transfer performance on electronic materials which are to be bonded with such a film adhesive.

In the present invention, spherical or amorphous fine particles may be used as the silicone particles, and fine particles with an average particle diameter of from 0.1  $\mu\text{m}$  to 20  $\mu\text{m}$  may preferably be used. Also preferred are silicone particles in which particles not larger than the average particle diameter hold 80% by weight or more of the particle size distribution of the fine particles. Silicone particles the particle surfaces of which have been treated with a silane coupling agent are particularly preferred because the dispersibility in the resin is improved.

The silicone particles used in the adhesive of the present invention may preferably have a modulus of elasticity of from 0.1 to 100 MPa at room temperature (25°C), and more preferably from 1 to 30 MPa in order to improve the dispersibility of the particles or to lessen the interfacial stress acting at the time of connection. Incidentally, the modulus of elasticity as defined herein is the modulus of elasticity of silicone rubber obtained by polymerizing the silane compound (such as an alkoxy silane or a partial hydrolysis-condensation product thereof) which is a material for the silicone

particles, and is measured by dynamic extended viscoelastometry.

The silicone particles may directly be mixed in the radically polymerizable substance, the curing agent 5 capable of generating a free radical upon heating or the film-forming material. Since, however, the former is readily dispersible in the film-forming material or radically polymerizable substance, the former may preferably be mixed with the latter after the former 10 has been dispersed in an organic solvent.

The silicone particles may be mixed in an amount of from 5 to 200 parts by weight, and more preferably from 10 to 50 parts by weight, based on 100 parts by weight of the total weight of the radically polymerizable 15 substance(s) (when the film-forming material is contained, based on 100 parts by weight of the total of the radically polymerizable substance(s) and the film-forming material). If the silicone particles are less than 5 parts by weight, the good bond strength to the substrate 20 which supports connecting terminals and to the wiring member surface and the releasability from the base material may be less achievable. If on the other hand the silicone particles are more than 200 parts by weight, the adhesive may have a low cohesive force and hence there is a

possibility that any good adhesive can not be obtained.

(4) Film-forming material:

Film-forming materials preferable for the present invention may include polyvinyl formal resin, polystyrene 5 resin, polyvinyl butyral resin, polyester resin, polyamide resin, xylene resin, phenoxy resin and polyurethane resin.

The film-forming material refers to a material which can make the adhesive be handled as a film in a usual 10 condition when a liquid composition is solidified into a film, i.e., a material which imparts good mechanical properties as film (the properties that the film formed can be handled with ease and the film does not split, break or become tacky) to the film to be formed. In view 15 of the readiness to handle as a film, a material capable of forming a self-supporting film is preferred.

Among compounds which can impart such properties, it is preferable to use phenoxy resin, as having superior 20 adhesion, compatibility, heat resistance and mechanical strength. The phenoxy resin is obtained by allowing a bifunctional phenol to react with an epihalohydrin to a product with a high molecular weight, or by subjecting a bifunctional epoxy resin and a bifunctional phenol to polyaddition.

Stated specifically, it can be obtained by, e.g.,  
allowing 1 mol of a bifunctional phenol to react with  
0.985 to 1.015 mol of an epihalohydrin in the presence  
of an alkali metal hydroxide in an inactive solvent at  
5 40 to 120°C.

From the viewpoint of the mechanical properties and thermal properties of the resin, particularly preferred is a product obtained by subjecting to polyaddition reaction a bifunctional epoxy resin and a bifunctional phenol in a mixing equivalent ratio of epoxy group/phenol hydroxyl group = 1/0.9 to 1/1.1, in the presence of a catalyst such as an alkali metal compound, an organic phosphorus compound or a cyclic amine compound, in an organic solvent of an amide type, an ether type, a ketone type, a lactone type or an alcohol type, having a boiling point of 120°C or above, in a reaction solid content of 50 parts by weight or less, and with heating at 50 to 200°C.

The bifunctional epoxy resin may include bisphenol A epoxy resin, bisphenol F epoxy resin, bisphenol AD epoxy resin and bisphenol S epoxy resin. The bifunctional phenol is a compound having two phenolic hydroxyl groups, and may include, e.g., hydroquinones and bisphenols such as bisphenol A, bisphenol F, bisphenol AD and bisphenol

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S. The phenoxy resin may be modified with a radically polymerizable functional group.

(5) Conductive particles:

The adhesive of the present invention need not particularly contain conductive particles because the electrical conduction is attained by direct contact of the wiring terminals to be connected. The conductive particles may preferably be contained because more stable connection can be attained.

Conductive particles preferable for the present invention may include particles of metals such as Au, Ag, Ni, Cu and solder, and carbon particles. In order to achieve a sufficient pot life, it is preferable that not a transition metal such as Ni or Cu but a noble metal such as Au, Ag or a platinum group metal forms their surface layers. It is particularly preferable that the surface is formed of Au. Particles comprised of a transition metal such as Ni the particle surfaces of which have been coated with a noble metal such as Au are also preferable for the present invention.

Composite particles comprised of non-conductive glass, ceramic or plastic particles on the surfaces of which conductive layers formed of the above metal have been formed to provide outermost layers formed of a noble

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metal, and heat-fusible metal particles are also suited for the present invention because they are deformable upon application of heat and pressure and hence the area of their contact with electrodes increases at the time 5 of connection to bring about an improvement in reliability.

In the case when the composite particles having coat layers of a noble metal at the surfaces are used, the coat layers may preferably be provided in a thickness of 100 angstroms or more in order to attain a good resistance. 10 Especially when the layers of a noble metal are provided on the transition metal such as Ni, the coat layers formed of a noble metal may preferably be provided in a thickness of 300 angstroms or more in order to prevent free radicals from being generated by the redox reaction caused by 15 any defects of the noble metal layers which defects may occur when the conductive particles are mixed and dispersed, to cause a lowering of storage stability. If, however, the coat layers are provided in a thickness larger than 1  $\mu\text{m}$ , the effect is no longer improved in proportion 20 to the thickness. Hence, in usual cases, it is preferable for the coat layers to be in a thickness of 1  $\mu\text{m}$  or less, to which, however, the present invention is by no means limited.

The conductive particles may properly be used

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according to purpose, in the range of from 0.1 to 30 parts by volume based on 100 parts by volume of the resin component of the adhesive. In order to prevent adjoining wirings from being short-circuited by any excess 5 conductive particles, the conductive particles may more preferably be used in an amount of from 0.1 to 10 parts by volume.

Structure in which the conductive particles are not in contact with the curing agent enables more improvement 10 of pot life. More specifically, an adhesive film having a double or more multi-layer structure in which at least a first layer formed of the adhesive of the present invention and a second layer formed of an adhesive mixed with the conductive particles in place of the curing 15 agent are formed in layers is preferred because a much longer pot life can be achieved.

(6) Other additives:

In the adhesive of the present invention, allyl acrylate and/or allyl methacrylate may optionally be 20 mixed in order to improve bond strength. It or they may be mixed in an amount of from 0.1 to 10 parts by weight, and more preferably from 0.5 to 5 parts by weight, based on 100 parts by weight of the total weight of the radically polymerizable substance(s) (when the film-forming

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material is contained, based on 100 parts by weight of the total of the radically polymerizable substance(s) and the film-forming material). If it is less than 0.1 part by weight, the effect of improving bond strength 5 may not sufficiently be achieved. If it is more than 10 parts by weight, the radical polymerization reaction may be so low as to cause insufficient reaction to make it difficult to achieve good bond strength.

In the adhesive of the present invention, a polymer 10 or copolymer having as a monomer component at least one of acrylic acid, an acrylate, a methacrylate and acrylonitrile may also be mixed. In particular, a copolymer acrylic rubber containing glycidyl acrylate and/or glycidyl methacrylate monomer(s) having a glycidyl 15 ether group promises superior stress relaxation, and may preferably be contained. Such an acrylic rubber may preferably have a weight-average molecular weight of 200,000 or more in order to make the adhesive have a higher cohesive force.

20 In the adhesive of the present invention, a filler, a softening agent, an accelerator, an anti-aging agents, a colorant, a flame retardant, a thixotropic agent, a coupling agent, a resin (e.g., phenolic resin or melamine resin) and an isocyanate may further be mixed.

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The mixing of the filler is preferable because the connection reliability and so forth are improved. In the case when the filler is used, the maximum diameter of its particles should be smaller than the particle 5 diameter of the conductive particles. Also, it may be mixed in an amount of from 5 to 60 parts by volume based on 100 parts by volume of the resin component in the adhesive. If it is more than 60 parts by volume, the effect of improving reliability may be saturated. If 10 it is less than 5 parts by volume, its addition is less effective.

As the coupling agent, any of ketimine-, vinyl-group-, acrylic-group-, amino-group-, epoxy-group- and isocyanate-group-containing agents are preferable for 15 the present invention in view of an improvement in adhesion.

As a silane coupling agent having an amino group, it may include, e.g.,  
N- $\beta$ -(aminoethyl) $\gamma$ -aminopropyltrimethoxysilane,  
20 N- $\beta$ -(aminoethyl) $\gamma$ -aminopropylmethyldimethoxysilane,  
 $\gamma$ -aminopropyltriethoxysilane,  
N-phenyl- $\gamma$ -aminopropyltrimethoxysilane and the like.

As a silane coupling agent having a ketimine, it may include those obtained by allowing the above silane

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coupling agent having an amino group, to react with a ketone compound such as acetone, methyl ethyl ketone or methyl isobutyl ketone.

(7) Uses:

5        The adhesive of the present invention may also be used as an adhesive film used to bond IC (integrated circuit) chips to chip-mount substrates or to bond electric wirings to one another. More specifically, the adhesive of the present invention which has been shaped into a 10 film (the adhesive film) may be interposed between a first wiring member having a first connecting terminal and a second wiring member having a second connecting terminal, and these may be heated and pressed to connect the first connecting terminal and the second connecting 15 terminal electrically.

Wiring members preferable as members to be connected according to the present invention may include chip component parts such as semiconductor chips, resistor chips and capacitor chips, and substrate members such 20 as printed-wiring substrates. These wiring members are provided with connecting terminals usually in a large number (may be provided with one terminal in some cases).

At least one set of such wiring members is so disposed that at least part of wiring terminals provided thereon

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face to one another, interposing the adhesive of the present invention between these, which are then heated under application of a pressure to electrically interconnect the connecting terminals facing to one another. Thus, a wiring structure (such as a wiring board) having two or more wiring members can be manufactured.

5 Here, the electrical conduction between wiring terminals thus formed may be set up by direct contact between the wiring terminals or may be set up via the conductive 10 particles contained in the adhesive.

The connection of wiring terminals according to the present invention may be performed by, e.g., forming an adhesive layer on the surface of the first connecting terminal (circuit electrode), and disposing on the surface 15 of this adhesive layer the second connecting terminal (circuit electrode) under registration in such a way that the connecting terminals face to each other, followed by heating and pressing. The adhesive layer may be formed by, e.g., coating a liquid adhesive, or may be formed 20 by placing the adhesive film.

(8) Physical properties of adhesive:

The adhesive of the present invention melts and flows at the time of connection to connect wiring terminals facing to one another, and thereafter it is cured to

keep the connection. Hence, the flowability of the adhesive is an important factor. The adhesive of the present invention may preferably have a flowability of from 1.3 to 3.0, and more preferably from 1.5 to 2.5, 5 as the value of flowability (B)/(A) represented by the initial area (A) and the area (B) after heating and pressing when the adhesive of the present invention which is of 35  $\mu\text{m}$  in thickness and 5 mm  $\times$  5 mm in size is interposed between two sheets of glass of 0.7 mm in thickness and 10 15 mm  $\times$  15 mm in size and these are heated and pressed at 150°C and 2 MPa for 10 seconds. If the value is smaller than 1.3, the adhesive may have so poor a flowability as not to achieve any good connection. If it is greater than 3.0, air bubbles may occur to result in a poor 15 reliability.

The adhesive of the present invention may also preferably have a modulus of elasticity of from 100 to 3,000 MPa at 25°C after curing, and more preferably from 300 to 2,000 MPa. When it has the modulus of elasticity 20 in this range, the resin can have a low internal stress after connection. Hence, this is advantageous for the improvement of adhesive force, and also can ensure good conduction performance.

The adhesive of the present invention may preferably

have, in measurement with a differential scanning calorimeter (DSC) at a heating rate of 10°C/minute, an exotherm rise temperature (Ta) of from 70°C to 110°C, a peak temperature (Tp) of Ta + 5 to 30°C and an end 5 temperature (Te) of 160°C or below.

## EXAMPLES

#### A. Preparation of adhesive:

### Example 1

10 (1) Synthesis of urethane acrylate:

400 parts by weight of polycaprolactone diol with an average molecular weight of 800, 131 parts by weight of 2-hydroxypropyl acrylate, 0.5 part by weight of dibutyltin dilaurate as a catalyst and 1.0 part by weight of hydroquinone monomethyl ether as a polymerization inhibitor were heated to 50°C with stirring to mix them. Next, 222 parts by weight of isophorone diisocyanate was dropwise added thereto, and the mixture formed was further heated to 80°C with stirring to effect urethanation reaction. Having made sure that the conversion of isocyanate groups reached 99% or more, the reaction temperature was dropped to obtain urethane acrylate A.

## (2) Synthesis of silicone particles:

Silicone particles were obtained by adding

methyltrimethoxysilane to an aqueous alcohol solution of pH 12 and kept at 20°C, with stirring at 300 rpm to effect hydrolysis and condensation. The silicone particles thus obtained had a modulus of elasticity of 5 8 MPa at 25°C and an average particle diameter of 2  $\mu\text{m}$ .

(3) Preparation of conductive particles:

Conductive particles were prepared by providing nickel layers of 0.2  $\mu\text{m}$  thick on the surfaces of particles composed of polystyrene as nuclei, and providing gold 10 layers of 0.04  $\mu\text{m}$  thick on the outsides of the nickel layers. The conductive particles thus obtained had an average particle diameter of 10  $\mu\text{m}$ .

(4) Preparation of adhesive:

100 parts by weight of the silicone particles obtained 15 in the step (2) were dispersed in 100 parts by weight of a mixed solvent of toluene/ethyl acetate of 50/50 in weight ratio.

The urethane acrylate A obtained in the step (1), a phosphate type acrylate (available from Kyoeishayushi 20 Kabushiki Kaisya; trade name: P2M), the silicone particles and t-hexyl peroxy-2-ethylhexanone (a free radical generator) were so mixed as to be in amounts of 99 g, 1 g, 30 g and 5 g, respectively, in solid weight ratio, and 3% by volume of the conductive particles obtained

in the step (3) were dispersed to obtain a liquid adhesive.

Example 2

50 g of phenoxy resin (available from Union Carbide Corporation; trade name: PKHC; average molecular weight: 5 45,000) was dissolved in a mixed solvent of toluene (boiling point: 110.6°C; SP value: 8.90)/ethyl acetate (boiling point: 77.1°C; SP value: 9.10) of 50/50 in weight ratio to form a solution with a solid content of 40% by weight.

10 The phenoxy resin, the urethane acrylate A, the phosphate type acrylate, the t-hexyl peroxy-2-ethylhexanone and the silicone particles were so mixed as to be in amounts of 50 g, 49 g, 1 g, 5 g and 5 g, respectively, in solid weight ratio, and 3% 15 by volume of the conductive particles were dispersed.

The dispersion thus obtained was coated on a one-side surface-treated PET (polyethylene terephthalate) film of 80 µm thick by means of a coater, followed by drying with 70°C hot air for 10 minutes to obtain an adhesive 20 film of 20 µm thick.

Examples 3 to 5

Adhesive films were obtained in the same manner as in Example 2 except that the solid weight ratio of phenoxy resin/urethane acrylate A/phosphate type

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acrylate/silicone particles/t-hexyl

peroxy-2-ethylhexanone was changed to 50 g/49 g/1 g/20 g/5 g (Example 3), 30 g/69 g/1 g/10 g/5 g (Example 4) and 30 g/40 g/30 g/10 g/5 g (Example 5).

5       Comparative Example 1

An adhesive film was obtained in the same manner as in the step (4) of Example 1 except that the solid weight ratio of urethane acrylate A/phosphate type acrylate/t-hexyl peroxy-2-ethylhexanone was changed 10 to 99 g/1 g/5 g and the silicone particles were not mixed.

Comparative Example 2

An adhesive film was obtained in the same manner as in Example 2 except that the silicone particles were not used.

15       B. Evaluation of adhesive:

(1) Production of wiring structure:

First, on the surface of a glass substrate 11 of 1.1 mm thick, wirings 12 of indium-tin oxide (ITO) were formed by vacuum deposition to prepare an ITO substrate 20 member (surface resistivity: <20  $\Omega/\text{square}$ ) 10 [Fig. 1 (a)]. On its side where the wiring 12 was formed, an adhesive layer 15 formed of an adhesive containing conductive particles 14 (the one prepared in each Example and Comparative Example) was formed [Fig. 1 (b)].

In Example 1 and Comparative Example 1, in which the adhesive was liquid, the adhesive layer 15 was formed by coating the adhesive. In Examples 2 to 5 and Comparative Example 2, in which the adhesive was filmy, it was formed 5 by fastening the adhesive to the wiring member, heating and pressing them at 70°C and 0.5 MPa for 5 seconds to effect provisional connection, and thereafter peeling the PET film off.

On the surface of this adhesive layer 15, a flexible 10 wiring board (a triple-layer FPC) was placed which was prepared by bonding a polyimide layer 18 and copper foil (thickness: 18  $\mu\text{m}$ ) with an adhesive 17 and subjecting the copper foil to patterning to form wirings 16 of 50  $\mu\text{m}$  in line width and 100  $\mu\text{m}$  in pitch, and these were 15 heated and pressed at 160°C and 3 MPa for 10 seconds to make connection over a width of 2 mm. Thus, a wiring structure 21 shown in Fig. 1 (d) was obtained.

A wiring structure 25 was also obtained [Fig. 2 (b)] by making connection to the ITO substrate member in the 20 same way but using, in place of the triple-layer FPC, a flexible wiring board (a double-layer FPC) 24 prepared by forming on the surface of a polyimide film (thickness: 100  $\mu\text{m}$ ) 22 a copper circuit 23 with 500 wirings of 50  $\mu\text{m}$  in line width, 100  $\mu\text{m}$  in pitch and 18  $\mu\text{m}$  in thickness [Fig.

2 (a)].

(2) Measurement of connection resistance:

After the wiring structures were produced in the manner as described above, their resistance value between 5 adjoining circuits of the FPCs including wiring-connected portions was measured with a multi-meter immediately after the production. These were further kept in a high-temperature high-humidity chamber of 85°C and 85%RH for 500 hours, and thereafter the resistance value was 10 measured in the same way. Here, the resistance value was shown as the average of 150 points of resistance between adjoining circuits.

(3) Measurement of bond strength:

The wiring structures produced in the manner as 15 described above were put to a peel test of peeling at 90 degrees at a peel rate of 50 mm/minute, to measure their bond strength.

(4) Evaluation of insulation properties:

First, a printed-wiring substrate having a 20 comb-shaped circuit in which a copper circuit with 250 wirings provided alternately in a line width of 50  $\mu\text{m}$ , a pitch of 100  $\mu\text{m}$  and a thickness of 18  $\mu\text{m}$  was prepared. On its side where the circuit was formed, an adhesive layer was formed in the same manner as the above (1).

Next, on the surface of this adhesive layer, a flexible wiring board (FPC) having a copper circuit with 500 wirings of 50  $\mu\text{m}$  in line width, 100  $\mu\text{m}$  in pitch and 18  $\mu\text{m}$  in thickness was placed, and these were heated and pressed at 160°C and 3 MPa for 10 seconds to make connection over a width of 2 mm. Thus, a wiring structure was obtained.

To the comb-shaped circuit of this wiring structure, a voltage of 100 V was applied to measure insulation resistance value. The wiring structure was further put to a high-temperature high-humidity test of 85°C and 85%RH for 500 hours, and thereafter the insulation resistance value was measured.

#### (5) Evaluation of flowability:

An evaluation-purpose adhesive of 5 mm x 5 mm in size and 35  $\mu\text{m}$  in thickness was sandwiched between two sheets of glass of 15 mm x 15 mm in size and 0.7 mm in thickness, and these were heated and pressed at 150°C and 2 MPa for 10 seconds, where the value of flowability (B) / (A) was determined on the basis of the initial area (A) and the area (B) after heating and pressing.

(6) Modulus of elasticity after curing:

The liquid adhesives (Example 1 and Comparative Example 1) were each casted into a mold, and then heated at 160°C for 1 minute to effect curing to obtain a rod-like

cured product. The filmy adhesives (Examples 2 to 5 and Comparative Example 2) were each immersed in 160°C oil for 1 minute to effect curing to obtain a film-like cured product. The storage elastic modulus of each of these 5 cured products was measured with a dynamic viscoelastometer (heating rate: 5°C/minute; 10 Hz) to determine the modulus of elasticity at 25°C.

(7) Measurement by DSC:

Using the adhesives obtained in the respective 10 Examples and Comparative Examples, the rise temperature (Ta), peak temperature (Tp) and end temperature (Te) of exothermic reaction were determined by means of a differential scanning calorimeter (DSC, manufactured by TA Instruments Co.; trade name: Model 910). In the 15 measurement, the heating rate was set at 10°C/minute.

C. Results:

The results obtained by the above evaluation methods are shown in Table 1.

Table 1

Adhesive composition			Example					Comparative Example		
			1	2	3	4	5	1	2	
Adhesive Force (N/m)	Double-layer FPC	Initial stage	400	700	750	650	680	50	100	
		After moisture absorption	200	400	550	450	400	10	20	
Connection resistance (Ω)	Triple-layer FPC	Initial stage	900	1200	1300	1100	1250	600	700	
		After moisture absorption	600	900	1000	1000	950	300	500	
Insulation resistance (Ω)			>10 <sup>9</sup>	>10 <sup>9</sup>						
Flowability (%)			2.5	1.9	1.7	1.8	1.9	2.6	1.8	
Modulus of elasticity (MPa)			500	800	700	800	800	600	700	
DSC (°C)		T <sub>a</sub>	88	87	90	89	89	87	88	
		T <sub>p</sub>	105	108	107	109	108	104	107	
		T <sub>e</sub>	140	148	147	147	148	143	148	

The silicone particles are not contained in the adhesives of Comparative Examples 1 and 2. In contrast thereto, in Examples 1 to 5, in which the silicone particles are mixed, the adhesive force is greatly improved compared with that in Comparative Examples, even in the case of the double-layer FPC in which the polyimide resin stands

uncovered to the surface of the substrate which supports connecting terminals and also in the case of the triple-layer FPC in which the adhesive stands uncovered thereto. The adhesive force after moisture absorption 5 is also great.

#### POSSIBILITY OF INDUSTRIAL APPLICATION

As described above, according to the present invention, a high bond strength can be achieved and a wiring structure having a high connection reliability 10 can be produced, even in the case of the wiring member in which the substrate which supports wiring terminals is made of an insulating organic matter such as polyimide resin, or glass, or in the case of the wiring member having silicon nitride, silicone resin and/or polyimide 15 resin at its surface.

1 **What is Claimed is:**

1 1. **A wiring-terminal-connecting adhesive comprising:**  
2       a curing agent capable of generating a free radical upon  
3 heating;  
4       a radically polymerizable substance; and  
5       silicone particles having a modulus of elasticity of from 0.1  
6 MPa to 100 MPa at 25°C and an average particle diameter of from  
7 0.1 µm to 20 µm.

1 2. **The wiring-terminal-connecting adhesive according to claim**  
2 **1, wherein said silicone particles are contained in an amount of**  
3 **from 5 parts by weight to 200 parts by weight based on 100 parts by**  
4 **weight of said radically polymerizable substance.**

1 3. **The wiring-terminal-connecting adhesive according to claim**  
2 **1, which further comprises a film-forming material.**

1 4. **The wiring-terminal-connecting adhesive according to claim**  
2 **3, wherein said film-forming material is a phenoxy resin.**

1 5. **The wiring-terminal-connecting adhesive according to claim**  
2 **3 or 4, wherein said silicone particles are contained in an amount of**  
3 **from 5 parts by weight to 200 parts by weight based on 100 parts by**

4 weight of the total of said radically polymerizable substance and  
5 said film-forming material.

1 6. (Cancelled)

1 7. The wiring-terminal-connecting adhesive according to any  
2 one of claims 1 to 5, which further comprises conductive particles.

1 8. A wiring-terminal-connecting adhesive film comprising:  
2 a first layer formed of a composition containing a curing  
3 agent capable of generating a free radical upon heating, a radically  
4 polymerizable substance and silicone particles having a modulus of  
5 elasticity of from 0.1 MPa to 100 MPa at 25°C and an average  
6 particle diameter of from 0.1  $\mu$ m to 20  $\mu$ m; and  
7 a second layer formed of a composition containing conductive  
8 particles, a radically polymerizable substance and silicone particles  
9 having a modulus of elasticity of from 0.1 MPa to 100 MPa at 25°C  
10 and an average particle diameter of from 0.1  $\mu$ m to 20  $\mu$ m, wherein  
11 the first and second layers being formed in layers.

1 9. A method of connecting wiring terminals, comprising  
2 interconnecting connecting terminals electrically with the  
3 wiring-terminal-connecting adhesive according to any one of claims  
4 1 to 5, 7 and 8, wherein at least two wiring members have the

5 connecting terminal individually.

1 10. The method of connecting wiring terminals according to claim  
2 9, wherein at least one of said connecting terminals has  
3 a surface which is formed of at least one selected from gold,  
4 silver, tin, a platinum group metal and indium-tin oxide.

1 11. The method of connecting wiring terminals according to claim  
2 9 or 10, wherein at least one of said wiring members has  
3 a substrate comprising at least one of an insulating organic  
4 material and glass.

1 12. The method of connecting wiring terminals according to any  
2 one of claims 9 to 11, wherein at least one of said wiring members  
3 has  
4 at least one selected from silicon nitride, silicone resin and  
5 polyimide resin, at its surface.

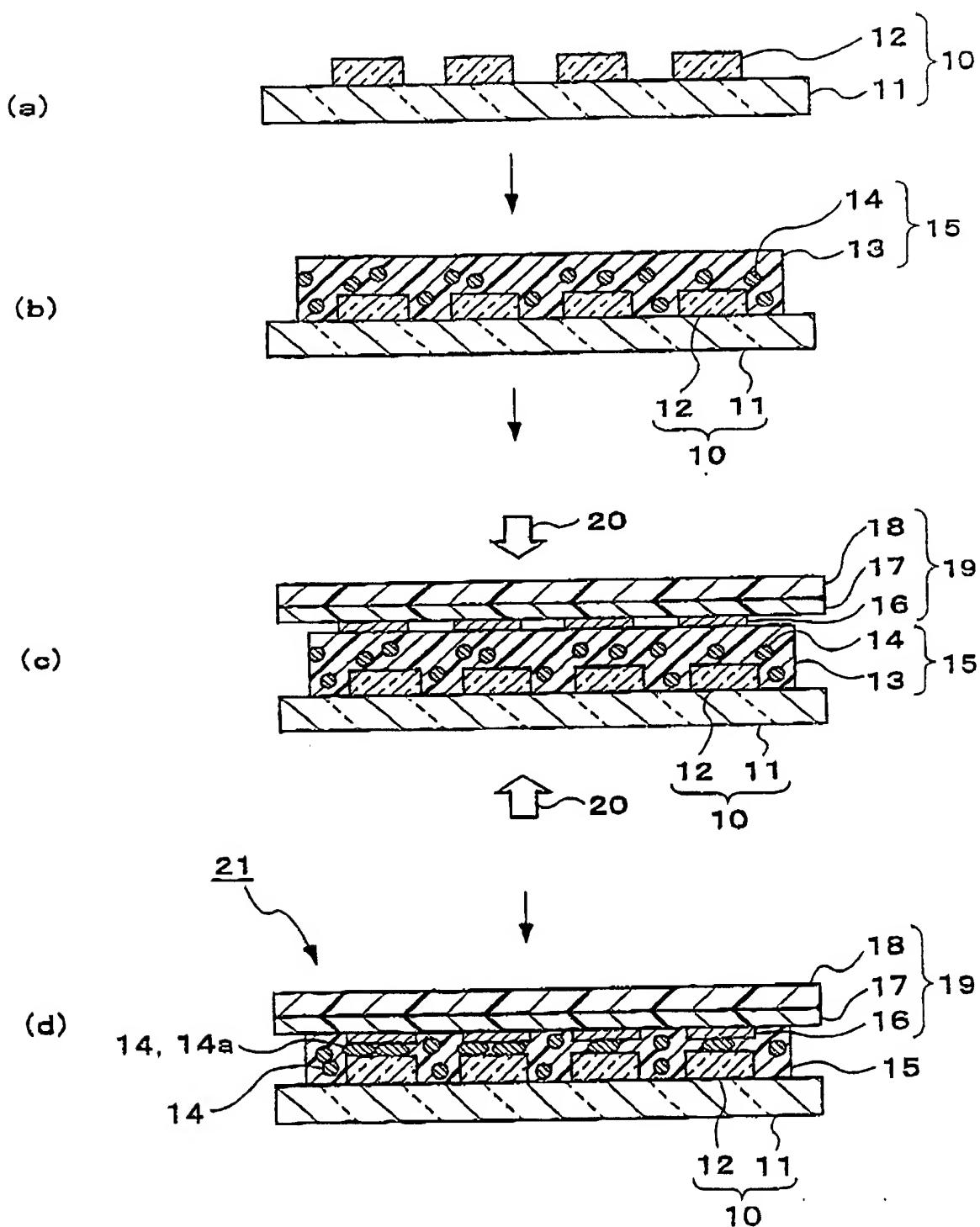
1 13. A wiring structure comprising at least two wiring members  
2 which have a connecting terminal individually, wherein  
3 the connecting terminals of said wiring members are  
4 electrically interconnected with the wiring-terminal-connecting  
5 adhesive according to any one of claims 1 to 5, 7 and 8.

ABSTRACT

This invention provides a wiring-terminal-connecting adhesive comprising a curing agent capable of generating a free radical upon heating, a radically polymerizable substance and silicone particles, and a wiring-terminal-connecting method and a wiring structure which make use of such an adhesive.

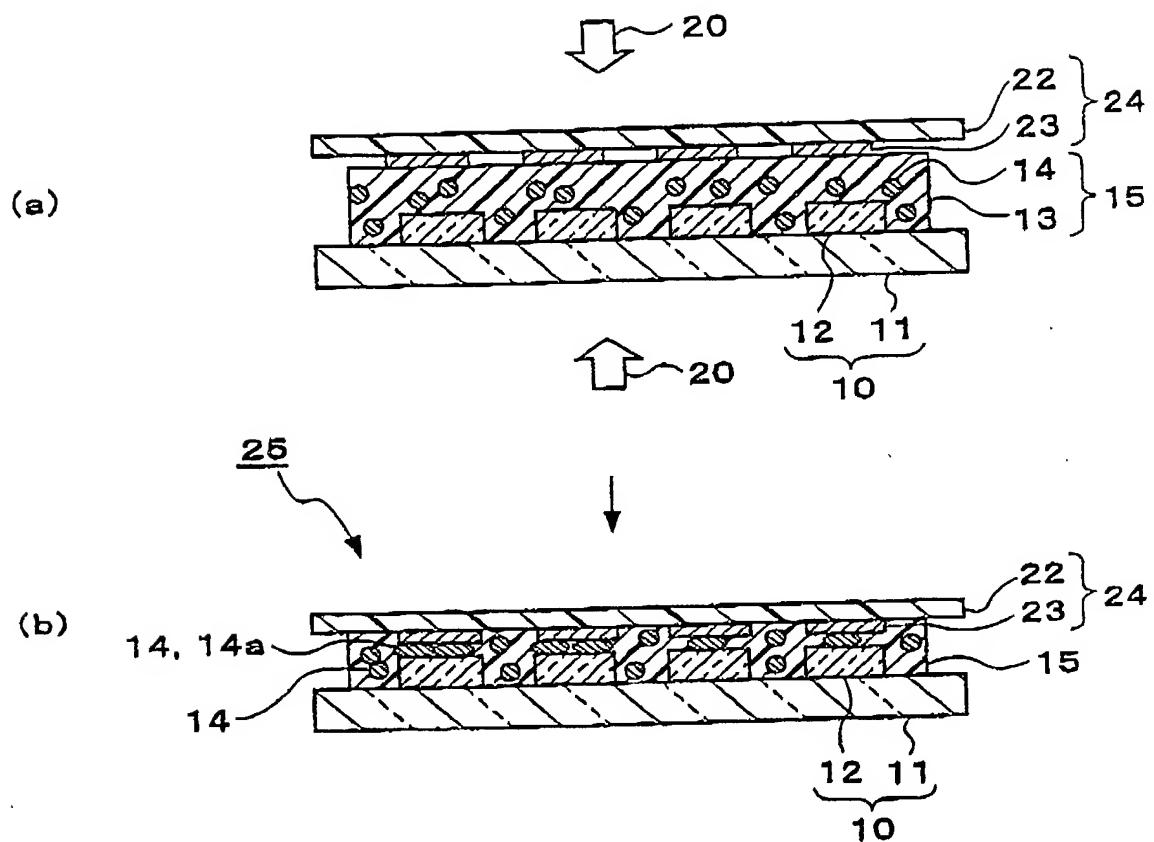
1/2

FIG. 1



2/2

FIG. 2



## Declaration and Power of Attorney for Patent Application

### 特許出願宣言書及び委任状

### Japanese Language Declaration

#### 日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。 As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

#### 接着剤、配線端子の接続方法及び配線構造体

ADHESIVE, METHOD OF CONNECTING WIRING

TERMINALS AND WIRING STRUCTURE

上記発明の明細書（下記の欄で印がついていない場合は、本書に添付）は、

2000年8月25日に提出され、米国出願番号または特許協定条約国際出願番号をPCT/JP00/05766とし、  
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the specification of which is attached hereto unless the following box is checked:

was filed on August 25, 2000  
as United States Application Number or PCT  
International Application Number  
PCT/JP00/05766 and was amended on  
February 25, 2002 (if applicable)

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

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私は、連邦規則法典第37編第1条56項に定義されるところ、特許資格の有無について重要な情報を開示する義務があることを認めます。

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Prior foreign application(s)  
外国での先行出願

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(Number)  
(番号)

JAPAN  
(Country)  
(国名)

\_\_\_\_\_  
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(国名)

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(Application No.)  
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(Filing Date)  
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(Application No.)  
(出願番号)

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Priority Not Claimed  
優先権主張なし

August 25, 1999  
(Day/Month/Year Filed)  
(出願年月日)

\_\_\_\_\_  
(Day/Month/Year Filed)  
(出願年月日)

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(Day/Month/Year Filed)  
(出願年月日)

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(出願年月日)

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(Day/Month/Year Filed)  
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I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application No.)  
(出願番号)

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I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Status)(patented, pending, abandoned)  
(現況：特許許可済、係属中、放棄済)

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

HC157603  
200024WUS0

Docket No. 566.41244X00

委任状： 私は、下記の発明者として、本出願に関する一切の手続を米国特許商標局に対して遂行する弁理士又は代理人として、下記のものを指名致します。（弁護士、又は代理人の氏名及び登録番号を明記のこと）

Donald R. Antonelli, Reg. No. 20,296; David T. Terry, Reg. No. 20,178; Melvin Kraus, Reg. No. 22,466; William I. Solomon, Reg. No. 28,565; Gregory E. Montone, Reg. No. 28,141; Ronald J. Shore, Reg. No. 28,577; Donald E. Stout, Reg. No. 26,422; Alan E. Schiavelli, Reg. No. 32,087; James N. Dresser, Reg. No. 22,973 and Carl I. Brundidge, Reg. No. 29,621

書類送付先：

Send Correspondence to:

Antonelli, Terry, Stout & Kraus, LLP  
Suite 1800  
1300 North Seventeenth Street  
Arlington, Virginia 22209

Direct Telephone Calls to:

Telephone: (703) 312-6600  
Fax: (703) 312-6666

唯一又は第一発明者名 有福 征宏	100	Full name of sole or first inventor <u>Motohiro ARIFUKU</u>
発明者署名	日付	Inventor's signature <u>Motohiro Arifuku</u> Date <u>2002. 5. 6.</u>
住所		Residence <u>Ibaraki, Japan</u> <u>JPX</u>
国籍 日本		Citizenship Japan
郵便の宛先 日立化成工業株式会社 五所宮事業所内 日本国茨城県下館市大字五所宮1150番地		Post Office Address c/o Goshomiya Works, Hitachi Chemical Company, Ltd. 1150, Oaza-Goshomiya, Shimodate-shi, Ibaraki 308-0866 Japan
第二共同発明者名（該当する場合） 渡辺 伊津夫	200	Full name of second joint inventor, if any <u>Itsuo WATANABE</u>
第二発明者署名	日付	Second inventor's signature <u>Itsuo Watanabe</u> Date <u>2002. 5. 6.</u>
住所		Residence <u>Ibaraki, Japan</u> <u>JPX</u>
国籍 日本		Citizenship Japan
郵便の宛先 日立化成工業株式会社 五所宮事業所内 日本国茨城県下館市大字五所宮1150番地		Post Office Address c/o Goshomiya Works, Hitachi Chemical Company, Ltd. 1150, Oaza-Goshomiya, Shimodate-shi, Ibaraki 308-0866 Japan

（第三以降の共同発明者についても同様に記載し、署名をすること。）

(Supply similar information and signature for third and subsequent joint inventors.)

第三共同発明者名（該当する場合） 本村 耕治		300	Full name of third joint inventor, if any <u>Kouji MOTOMURA</u>
第三発明者の署名		日付	Third inventor's signature Date <u>Kouji MOTOMURA</u> 2002.04.25
住所			Residence <u>Ibaraki, Japan JPX</u>
国籍 日本			Citizenship Japan
郵便の宛先 日立化成工業株式会社 五所宮事業所内 日本国茨城県下館市大字五所宮1150番地			Post office address c/o Goshomiya Works, Hitachi Chemical Company, Ltd.1150, Oaza-Goshomiya, Shimodate-shi, Ibaraki 308-0866 Japan
第四共同発明者名（該当する場合） 小林 宏治		400	Full name of fourth joint inventor, if any <u>Kouji KOBAYASHI</u>
第四発明者の署名		日付	Fourth inventor's signature Date <u>Kouji Kobayashi</u> 2002.05.06
住所			Residence <u>Ibaraki, Japan JPX</u>
国籍 日本			Citizenship Japan
郵便の宛先 日立化成工業株式会社 五所宮事業所内 日本国茨城県下館市大字五所宮1150番地			Post office address c/o Goshomiya Works, Hitachi Chemical Company, Ltd.1150, Oaza-Goshomiya, Shimodate-shi, Ibaraki 308-0866 Japan
第五共同発明者名（該当する場合） 後藤 泰史		500	Full name of fifth joint inventor, if any <u>Yasushi GOTOH</u>
第五発明者の署名		日付	Fifth inventor's signature Date <u>Yasushi Gotoh</u> 2002.05.06
住所			Residence <u>Ibaraki, Japan JPX</u>
国籍 日本			Citizenship Japan
郵便の宛先 日立化成工業株式会社 五所宮事業所内 日本国茨城県下館市大字五所宮1150番地			Post office address c/o Goshomiya Works, Hitachi Chemical Company, Ltd.1150, Oaza-Goshomiya, Shimodate-shi, Ibaraki 308-0866 Japan
第六共同発明者名（該当する場合） 藤繩 貢		600	Full name of sixth joint inventor, if any <u>Tohru FUJINAWA</u>
第六発明者の署名		日付	Sixth inventor's signature Date <u>Tohru Fujinawa</u> 2002.05.06
住所			Residence <u>Ibaraki, Japan JPX</u>
国籍 日本			Citizenship Japan
郵便の宛先 日立化成工業株式会社 総合研究所内 日本国茨城県つくば市和台48番			Post office address c/o Research & Development Center, HITACHI CHEMICAL CO., LTD. 48, Wadai, Tsukuba-shi, Ibaraki 300-4247 Japan

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